

SUB-WETTING LAYER CONTINUUM STATES IN QUANTUM DOT SAMPLES

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Abstract. In the polar semiconductor samples of the self-organized quantum dots, grown by the Stranski-Krastanow growth method, the lowest energy extended states of the electronic excitations are assumed to be the wetting-layer states. The coupling between these extended states and the electronic states localized in the individual quantum dots, may influence the optical spectra of such samples in the sub-wetting layer region of energy. This effect is studied assuming the Fröhlich's coupling between the electrons and polar optical phonons. The contribution of this interaction to the appearance of the sub-wetting layer continuum in the optical spectra and to the level broadening of the localized states, pointed out in some experiments, is estimated.

1. Introduction

The semiconductor nanoparticles are promising from the point of view of both basic science and applications [1]. One of their significant features is that the charge carriers can be localized within these nanoparticles. The quantum dots are thus regarded as a realization of artificial atoms with the nearly delta function-like spectral density of the electronic states. The quantum dot aggregates are prepared very often by the Stranski-Krastanow growth technique [1]. In the self-assembled quantum dot (SAQD) samples, prepared by this method, the quantum dots are grown on the top of the wetting layer (WL), with which the quantum dots interact. The optical absorption spectra of nanoparticles dispersed in a polymer film, which also seem to display an absorption continuum background increasing with the energy of the absorbed photon [2, 3], are not discussed here.

In the type *I* quantum dots [1] in the undoped SAQD samples, the lowest energy excitations are the states with the electrons excited over the semiconductor band gap to the conduction band electronic quantum dot localized states, leaving the holes in the valence-band states of the quantum dot. In the SAQD samples it is expected that the lowest energy electronic excitations, which are not localized in the quantum dots, are the excitations to the wetting layer states [4, 5]. The scheme of the electronic states available to the electronic excitations in the SAQD samples is drawn in the Fig. 1.

The electronic system can also be excited to the extended states belonging to the substrate and to the electronic states in the cladding layers [6]. With a certain simplification, the electronic states localized in the quantum dots can be expected to have a delta function like density of states. This may be seen in contrast with experiments [4, 6, 7, 8, 9, 10, 11, 12, 13, 14] showing that the spectral densities may be different from delta functions and that the width of them may depend on the intensity of laser light exciting the SAQD sample. Besides this observation, the experiments on luminescence and optical absorption suggest that in the sub-wetting layer energy region, where we expect only bound electronic states, a continuum background is observed.

The continuous background, which seems characteristic of the SAQD samples, occurs often in the measurements on samples with a rather high density of the quantum dots and a large density of electrons excited to the WL states [14]. The presence of high densities of charge carriers may lead us to consider the electrostatic coupling among the charge carriers [15, 16, 17]. We shall not consider this mechanism in the present work. Also, we shall neglect any direct coupling between quantum dots, like the electron tunneling [18], electrostatic coupling between the dots, or any other. Recently, the Fröhlich's coupling [19] between the electrons and dispersionless polar optical phonons has been applied to the interpretation of the energy relaxation of electrons in quantum dots [20, 21] and to other properties [20, 22, 23]. The theory of the energy relaxation in the two level system with the dispersionless phonons has recently been supported by an exact calculation [24]. The purpose of this work is to show the possible contribution of the Fröhlich's coupling to the origin of the sub-WL continuous background spectral density and to the electronic level broadening in the SAQD samples.

2. The theoretical model of the SAQD system

Trying to study the optical spectra in the rather complicated samples, as the SAQD are, we are led to make a number of simplifications, which make

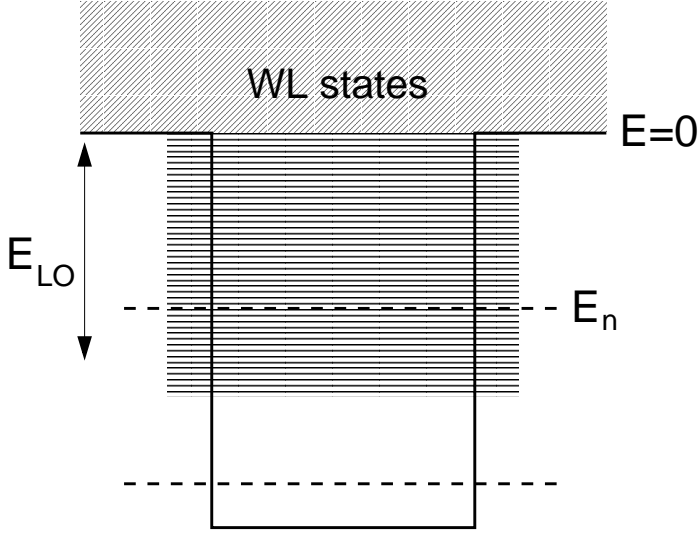


Figure 1. Schematic picture of the energetic structure of the single-electron states in the self-assembled quantum dot (SAQD) sample. The full line is the potential profile of the electrons in the quantum dot. The dashed horizontal lines denote the states bound in the quantum dots. The oblique hatched area denotes the extended electronic states in the wetting-layer. The horizontal hatching marks the sub-wetting layer area of electronic energies. E_n is the energy of a selected bound state in the sub-wetting layer energy region, E_{LO} is optical phonon energy.

the problem tractable, but which hopefully do not influence principally the main conclusions of the work. Dealing with the electron-phonon Fröhlich's interaction we shall neglect the influence of the interfaces in the SAQD heterostructure on the structure of the optical phonons. From the reasons given e. g. in the ref. [25] we shall simply assume that the charge carriers interact with the dispersionless longitudinal phonons of the bulk crystal of GaAs. Neglecting the effect of the electrostatic interaction among electrons and holes, we neglect also the exciton effect. We shall assume the limit of very heavy holes, in which their influence is reduced only to the static electrostatic field contributing to the effective potential in which the electrons move in the quantum dot potential in the conduction band.

In order that the details of the present model Hamiltonian are better understood, let us remind the principal features of the structure of the SAQD sample. With a certain simplification, in the Stranski-Krastanow method the surface of a substrate is first covered by a thin layer of the material, called wetting-layer, from which the quantum dots are to be grown. After depositing this layer the sample is left to obey a spontaneous process.

In this process the wetting layer changes its structure, namely, due to the difference in the lattice constant between the substrate material and that of the wetting layer, the wetting layer starts to become thinner and the material of the wetting layer forms small nanocrystals on the top of the wetting-layer. These small nanostructures may have sometimes a shape of a pyramid. Because of the shape and the size these small structures are called quantum dots. The process stops at some size of the nanocrystals and at some remaining thickness of the wetting-layer. The final state is a result of an energetic equilibrium in the whole system of the interatomic forces of the system. In order to prevent the mutual transfer of the material between the wetting layer and the quantum dots after the growth is ended, all the structure is covered from the top by a capping layer of a material, which can be similar to the substrate material. In this structure the electrons appear to be localized in the quantum dots. In order to overcome this localization they either have to gain an energy to transfer to the capping layer, or they need to get an energy sufficient for to transfer to the lowest energy states in the very thin wetting layer. Because of the small thickness of the wetting layer, the lowest energies in this structure are rather large. Usually however, the lowest energetic barrier for the electron to leave the quantum dot is that to the wetting layer. The density of the quantum dots, with which they are distributed on the surface of WL, depends on the technological conditions of the Stranski-Krastanow process.

The wetting-layer is a quasi-two dimensional quantum well nanostructure. In the SAQD sample the electrons of the quantum dots are expected to be coupled to electronic states of the wetting layer. The complete orthonormal set of the electronic states in the quantum dots and in the wetting layer depend on the details of the geometry of the SAQD sample. Realizing that besides the WL extended states there are also other extended states in the real sample, namely those associated with the substrate and capping layers [4], which may be energetically not very far from the wetting-layer states, and in order to simplify the theory, we shall assume that the wetting layer is a three-dimensional subsystem with the electronic states extended throughout the whole SAQD sample. The impact of this dimensionality assumption can be verified by recalculating the present results with a two-dimensional wetting layer assumption. This will be postponed to a further work. Our model, using the three-dimensional wetting-layer continuum, may therefore correspond better to the vertically coupled three-dimensional stacks of the quantum dots [26], or, to the Volmer-Weber island growth technology without the wetting layer (see e. g. [27]).

We shall approximate the electronic bound states in the individual dots by the electronic eigenstates in the three dimensional infinitely deep quantum wells. Although we shall assume that the density of the quantum dots

in the sample is rather high, namely $8 \times 10^{21} \text{ m}^{-3}$, which by the mean inter-dot distance corresponds to the usual two-dimensional density of quantum dots in the measured samples [13], we shall assume that the single electron states in the individual quantum dots are mutually orthogonal. In this work, confining ourselves to the basic estimates, we shall need to consider only one bound state per a single dot. In real samples, the individual quantum dots have only a finite potential depth. In our model we shall therefore assume that the single-electron spectrum in a quantum dot is shifted by a certain energy, so that the energy of the bound state under consideration is positioned in the sub-wetting layer region of electronic energy and we shall neglect the other bound states in the dots.

The electronic motion in the WL states can be approximated by plane waves, with the electronic energy $E_{\mathbf{k}} = \hbar^2 |\mathbf{k}|^2 / (2m)$, m being the electron conduction band energy in GaAs. The plane waves together with the bound states would not make an orthogonal set of states. The problem of nonorthogonality of the basis can be approximately avoided upon restricting the magnitude k of the electron wave vectors \mathbf{k} in the WL states to the values $k \ll k_m$. In this work we shall assume that the electronic wavelength corresponding to k_m is $1.5 * d$, d being the lateral size of the cubic quantum dot. We assume $d = 20 \text{ nm}$. In GaAs this assumption limits the WL electronic energy from above by about 24 meV .

The restriction put on the extent of the WL space of states will be utilized also in another context. We know little about the state of electronic statistical distribution in the course of the experiment, so that we use a simple assumption of the population of the WL states. Namely, we shall assume that the quantum mechanical electronic WL states are populated by electrons with a homogeneous density $\overline{N}_{\mathbf{k}}$, independent of the electron wave vector. This homogeneity assumption simplifies the theoretical treatment in a certain way. The overall density of the WL electrons is then obviously determined by $\overline{N}_{\mathbf{k}}$ and k_m .

In the above specified single-electron basis the full Hamiltonian consists of free electron Hamiltonian in the bound and WL states, H_e , free phonon Hamiltonian, H_{ph} , and the electron-phonon coupling H_1 between them:

$$\begin{aligned}
 H &= H_e + H_{ph}, \\
 H_e &= \sum_{i,n} E_n c_{i,n}^+ c_{i,n} + \sum_{\mathbf{k}} E_{\mathbf{k}} c_{\mathbf{k}}^+ c_{\mathbf{k}}, \\
 H_{ph} &= \sum_{\mathbf{q}} E_{LO} b_{\mathbf{q}}^+ b_{\mathbf{q}}, \\
 H_1 &= \sum_{\lambda,\mu,\mathbf{q}} A_q \Phi(\lambda,\mu,\mathbf{q}) (b_{\mathbf{q}} - b_{-\mathbf{q}}^+) c_{\lambda}^+ c_{\mu}, \tag{1}
 \end{aligned}$$

Here λ and μ are the indexes of single electron states, either (i, n) , or \mathbf{k} , where i is the quantum dot number and n is the electron orbital number. c and b are, respectively, electron and phonon annihilation operators, E_{LO} is phonon energy. The coupling constant is [19]

$$A_q = -ieq^{-1}[E_{LO}(\kappa_\infty^{-1} - \kappa_0^{-1})^{1/2}(2\varepsilon_0 V)^{-1/2}. \quad (2)$$

Here κ_∞ and κ_0 are, respectively, the high frequency and static dielectric constants, ε_0 is the permittivity of the free space, V is the volume of the sample and e is the electronic charge. The form-factor in (1)

$$\Phi(\lambda, \mu, \mathbf{q}) = \int d^3\mathbf{r} \psi_\mu^*(\mathbf{r}) e^{i\mathbf{q}\mathbf{r}} \psi_\lambda(\mathbf{r}). \quad (3)$$

modifies the Fröhlich's coupling to the case of quantum dot. Because the Fröhlich's coupling does not change the electron spin, we treat the electronic subsystems with the given spin separately. The functions ψ are the single electron orbitals in the SAQD sample. The terms of H_1 corresponding the Fröhlich's coupling between two bound states localized in different quantum dots will be considered as zero.

3. Spectral density

We shall calculate the electronic spectral density for the Hamiltonian specified above. This basic quantity will be used to compare the theoretical results with some experimental data in photoluminescence, photoluminescence excitation and optical absorption experiments. The electronic spectral density $\sigma_{i,n}(E)$, E being the energy variable, is related to the retarded Green's function $G_{i,n}(E)$ with help of the formula $\rho_{i,n}(E) = -\frac{1}{\pi} \text{Im} G_{i,n}(E)$. Similarly, spectral densities and the corresponding Green's functions $G_{\mathbf{k}}(E)$ are introduced for the wetting layer electronic states.

The electronic Green's function can be determined by the corresponding electronic self-energy, $M_{i,n}(E)$. Only the diagonal terms, in the electronic orbital quantum number n , of the Green's function and self-energies, will be considered. The reader is referred to the earlier works on the electron energy relaxation in individual quantum dots [21, 28, 29, 30]. The bound states electronic self-energy in the self-consistent Born approximation reads:

$$\begin{aligned} M_{i,n}(E) = & \sum_m \alpha_{n,m} \left\{ \frac{1 - N_{i,m} + \nu_{LO}}{E - E_m - E_{LO} - M_{i,m}(E - E_{LO})} \right. \\ & + \left. \frac{N_{i,m} + \nu_{LO}}{E - E_m + E_{LO} - M_{i,m}(E + E_{LO})} \right\} \\ & + \sum_{\mathbf{k}} \alpha_{n,\mathbf{k}} \left\{ \frac{1 - N_{\mathbf{k}} + \nu_{LO}}{E - E_{\mathbf{k}} - E_{LO} - M_{\mathbf{k}}(E - E_{LO})} \right. \end{aligned}$$

$$+ \frac{N_{\mathbf{k}} + \nu_{LO}}{E - E_{\mathbf{k}} + E_{LO} - M_{\mathbf{k}}(E + E_{LO})} \Big\}, \quad (4)$$

while for the wetting layer electrons we have:

$$\begin{aligned} M_{\mathbf{k}}(E) = & \sum_m \alpha_{\mathbf{k},\mathbf{k}'} \left\{ \frac{1 - N_{\mathbf{k}'} + \nu_{LO}}{E - E_{\mathbf{k}'} - E_{LO} - M_{\mathbf{k}'}(E - E_{LO})} \right. \\ & + \left. \frac{N_{\mathbf{k}'} + \nu_{LO}}{E - E_{\mathbf{k}'} + E_{LO} - M_{\mathbf{k}'}(E + E_{LO})} \right\} \\ & + \sum_{i,s} \alpha_{\mathbf{k},i,s} \left\{ \frac{1 - N_{i,s} + \nu_{LO}}{E - E_s - E_{LO} - M_{i,s}(E - E_{LO})} \right. \\ & + \left. \frac{N_{i,s} + \nu_{LO}}{E - E_s + E_{LO} - M_{i,s}(E + E_{LO})} \right\}. \end{aligned} \quad (5)$$

Here i is the number of quantum dot, while n and s are the numbers of electron bound state orbitals in the given dot. $N_{i,m}$ is the number of electrons in the m -th state if i -th quantum dot, $N_{\mathbf{k}}$ is the number of electrons in the \mathbf{k} -th state of the wetting-layer states and ν_{LO} is the population of the optical phonon states of the sample, given here by the Bose-Einstein distribution function. In these equations for the electronic self-energy, the constants α are generally given by

$$\alpha_{\lambda,\mu} = \sum_{\mathbf{q}} |A_q|^2 |\Phi(\mu, \lambda, \mathbf{q})|^2, \quad (6)$$

where the summation extends over all optical phonon wave vectors \mathbf{q} . The above equations for the electronic self-energy are self-consistent. Generally, they correspond to including an infinite number of terms of the self-energy expansion in the powers of H_1 . The use of the self-consistency, corresponding to the inclusion of the multiple-phonon scattering, proved important in the case of the electron-energy relaxation among the discrete energy levels of an electron in quantum dots [20, 21] and would be important when considering multiple bound states in a single dot, coupled via H_1 . Those terms in the equations (4,5), corresponding to the scattering between one bound state and one extended state, and those, corresponding to the scattering of the electron between two extended states, will be included in the bare Born approximation only.

Let us now specify the constants α in the three specific cases of the choice of the electronic states. In the case when both λ and μ denote the states localized in the same quantum dot, $\lambda = (i, n)$, $\mu = (i, m)$, we find that the corresponding constant $\alpha_{n,m} = \alpha_{m,n}$ in the equation (6) does not

depend on the quantum dot site index i . This is because the form-factor depends on the site index i in the following way:

$$\Phi((i, m), (i, n), \mathbf{q}) = e^{i\mathbf{q}\mathbf{r}} \Psi^{(irr)}(m, n, \mathbf{q}), \quad (7)$$

where the irreducible part of the form-factor is defined as

$$\Psi^{(irr)}(m, n, \mathbf{q}) = \int d^3\mathbf{r} \psi_{(0,n)}^*(\mathbf{r}) e^{i\mathbf{q}\mathbf{r}} \psi_{(0,m)}(\mathbf{r}). \quad (8)$$

Here the site index 0 means the position of the quantum dot at the origin of coordinates. We therefore drop out the site index i from the constant α completely for the coupling between the bound states, and we have:

$$\alpha_{nm} = \sum_{\mathbf{q}} |A_q|^2 |\Psi^{(irr)}(m, n, \mathbf{q})|^2. \quad (9)$$

The constant $\alpha_{n,\mathbf{k}} = \alpha_{\mathbf{k},n}$, which characterizes the mutual coupling of the bound and extended states, is determined with help of the form-factor

$$\Phi((i, n), \mathbf{k}, \mathbf{q}) = V^{-1/2} \int d^3\mathbf{r} e^{i\mathbf{k}\mathbf{r}} e^{i\mathbf{q}\mathbf{r}} \psi_m(\mathbf{r}), \quad (10)$$

V is the volume of the sample. When the electron wave vector \mathbf{k} is put zero, the latter integral is the Fourier transform of the electronic wave function. Because this wave function is localized in the cube of the lateral dimension d , the Fourier transform will be a function of \mathbf{q} , the width of which in the \mathbf{q} -space will be about π/d . Having confined our space of available extended states to the plane waves the \mathbf{k} -vectors of which fulfill the condition $|\mathbf{k}| \ll \pi/d$, we can expect that in the limit of \mathbf{k} -vector going to zero, the magnitude of \mathbf{k} in the form-factor $\Phi((i, n), \mathbf{k}, \mathbf{q})$ will not have any important influence on the value of the corresponding constant α . In practice we will assume that $|\mathbf{k}| \leq k_m$, $2\pi/k_m = 1.5 \times d$. We get:

$$\alpha_{n,\mathbf{k}} \approx \alpha_{n,\mathbf{k}=0} = \sum_{\mathbf{q}} |A_q|^2 |\Phi^{(irr)}(\mathbf{k}=0, n, \mathbf{q})|^2, \quad (11)$$

where

$$\Phi^{(irr)}(\mathbf{k}=0, n, \mathbf{q}) = V^{-1/2} \int d^3\mathbf{r} e^{i\mathbf{q}\mathbf{r}} \psi_{0,n}(\mathbf{r}). \quad (12)$$

It is then straightforward to get $\alpha_{n,\mathbf{k}} = B_n/V$, where the constant B_n is

$$B_n = \frac{e^2 E_{LO} (\kappa_\infty^{-1} - \kappa_0^{-1})}{16\pi^3 \varepsilon_0} \left| \int d^3\mathbf{q} e^{i\mathbf{q}\mathbf{r}} \psi_{0n}(\mathbf{r}) \right|^2. \quad (13)$$

In the equation (5), the term which introduces the coupling between the extended states, is (we just rewrite the original Fröhlich's coupling)

$$\alpha_{\mathbf{k},\mathbf{k}'} \equiv \sum_{\mathbf{q}} |A_q|^2 |\Phi(\mathbf{k},\mathbf{k}',\mathbf{q})|^2, \quad (14)$$

with $\Phi(\mathbf{k},\mathbf{k}',\mathbf{q}) = \delta_{\mathbf{k},\mathbf{k}'+\mathbf{q}}$. Then $\alpha_{\mathbf{k}',\mathbf{k}} = C_{\mathbf{k}'-\mathbf{k}}/V$, and

$$C_{\mathbf{k}'-\mathbf{k}} = \frac{e^2 E_{LO} (\kappa_\infty^{-1} - \kappa_0^{-1})}{2\varepsilon_0 |\mathbf{k}' - \mathbf{k}|^2} = \frac{p}{|\mathbf{k}' - \mathbf{k}|^2}, \quad (15)$$

defining the quantity p by this formula.

4. The influence of the wetting layer states

We shall now consider the influence of the wetting layer states on the spectral density in the sub-wetting layer region of electronic energy. We have set the bottom of the wetting layer states at the energy $E = 0$. The electronic states localized in quantum dots are then at $E < 0$.

Inspecting the equations (4) and (5) we see that only some of the terms at the right hand side give singularities of the self-energy in the sub-wetting layer region, in the lowest order of iteration of these equations. These terms are only the following ones: the term corresponding to the last fraction in equation (4) and similarly the term containing the second fraction in equation (5). These terms bring phonon satellites into the electronic spectral density at the sub-WL region. Also, these terms give a contribution, which is proportional to the electronic population of the wetting-layer states, which property appears to be connected with the presence of the discrete level broadening in the sub-WL region [14].

The experiments, with which we wish to compare our results, are usually performed at 10 K. Therefore, we take $\nu_{LO} = 0$ for the population of the phonons. So that, unless the temperature of the lattice optical phonons is increased substantially, the above self-energy terms are nonzero only when there is a nonzero population of the wetting layer electron states. In the present numerical estimates we shall assume a rather large population in the wetting-layer states. The mechanism of achieving such a WL population is not discussed here in any detail. The large population of the WL states corresponds to the numerical estimates in the experimental papers [14].

4.1. A SINGLE BOUND STATE

Let us see the influence of the wetting layer states on the spectral density of a single bound electronic state. Let us assume the simple case, when there is only a single bound state in every quantum dot, which we denote as the

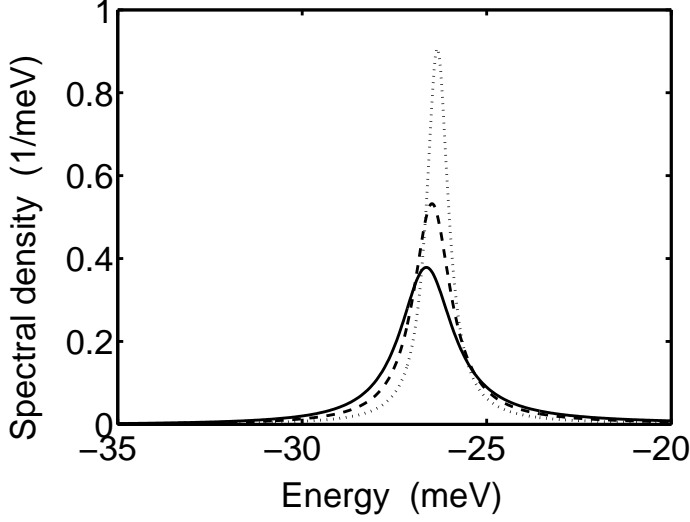


Figure 2. Spectral density $\sigma_n(E)$ of the bound state of a single quantum dot interacting with the wetting-layer states at the WL polulation: $\overline{N_{\mathbf{k}}} = 0.25$ (dotted line), 0.5 (dashed) and 0.75 (full).

state $n = 2$, the energy of which will be denoted as E_2 and $E_2 = -E_{LO} + \eta$. The parameter η determining the position of the bound state energy level is chosen to be 10 meV. In this work we use the material parameters of Gallium Arsenide. Namely, the electron relative effective mass is 0.067, the static dielectric constant is $\kappa_0 = 12.91$ (SI units) and the high frequency dielectric constant is $\kappa_\infty = 10.91$. The bulk crystal phonon energy in GaAs is 36.2 meV. The wave function of this state will be assumed to be that of an electron in one of the states of the triply degenerate first excited energy level of an infinitely deep cubic quantum dot with the lateral size d . Namely, this wave function is

$$\psi_{02}(\mathbf{r}) = (2/d)^{3/2} \cos(\pi x/d) \cos(\pi y/d) \sin(2\pi z/d) \quad (16)$$

(reminding that the index 0 means the wave function placed at the origin of coordinates). The natural line width of the spectral density of the bound state is taken here to be $\Delta = 0.05$ meV. The same natural line-width is assumed for the states in the wetting-layer. This means that the spectral density of the unperturbed bound state would be a Lorentzian, having the halfwidth of 0.05 meV, being placed at the energy $-E_{LO} + 10$ meV. The electronic self-energy term, expressed in the Born approximation to the coupling between the bound state ψ_{02} and the WL states, and being

proportional to the WL population at low temperatures, is simply

$$M_n(E) = \sum_{\mathbf{k}} \alpha_{n,\mathbf{k}} \frac{N_{\mathbf{k}}}{E - E_{\mathbf{k}} + E_{LO} + i\Delta}. \quad (17)$$

We omit the index of the quantum dot here. As said above, we assume here that the population of the WL states $N_{\mathbf{k}}$ is constant for all $\overline{N_{\mathbf{k}}}$. Under the above made assumption, according to which the wetting-layer states \mathbf{k} -vector magnitude is restricted to be not larger than k_m , this means that the average density of the electrons (of both spins) is about $1.5 \times 10^{23} \text{ m}^{-3}$ for $\overline{N_{\mathbf{k}}} = 0.5$. We assume that the electrons occupy only the WL states.

Reminding the above introduced approximation, according to which the wavevector dependence of the constant $\alpha_{n,\mathbf{k}}$ is neglected, the integral in the equation (17) can be integrated analytically [31]. The result of the numerical evaluation of the electronic spectral density $\sigma_n(E)$ of the electronic state $n = 2$, bound in a quantum dot, is presented in Fig. 2 for three values of the population of the wetting layer states. The spectral peak of the bound state is broadened, from the unperturbed value of the full width at half maximum being 0.1 meV, to the value of about 2 meV. This spectral width is of the same order of magnitude as it may be found in the experimental papers (see e. g. [14]). The Figure 2 shows the trend of the discrete state spectral density to broaden with the increase of the wetting layer states population. This behaviour of the bound state line-width appears to be in accord with the experiment [14]. The purpose of the present study is to pay attention to the main features of the effects under consideration. From this reason we use the material parameters of GaAs, as a typical material among the polar semiconductors. The quantum dot samples studied in experiment are often the InGaAs quantum dots, and the corresponding wetting layer, both grown on the substrate of GaAs. Although the parameters characterizing InGaAs are rather similar to those of GaAs, we nevertheless do not compare our results quantitatively with the experiment. In the case of the bound state energy level broadening, the reader is referred to the paper [14], namely to the Figure 3 in that paper, for the experimental data on the level broadening as it depends indirectly on the WL electron density. Let us emphasize that in the experimental works, with which we compare our results, the optical spectra are obtained in such a way, that they are measured in the limit of a single dot measurement and therefore the inhomogeneous broadening of the optical spectra is avoided. In this sense, we can therefore compare the experimental data with our "homogeneously" broadened theoretical shape.

Besides the broadening of the discrete electronic level, the coupling of the bound state to the WL states tends to give a weak continuous background. Quantitatively, this background appears rather weak at the presently used values of the input parameters of the material, quantum dot

size and shape, and at the present assumption about the populated states in the wetting-layer continuum. We may conclude that, the interaction of the bound state in the dot with the wetting layer states, under rather high population per quantum mechanical state, leads mainly to the bound state level broadening in the sub-wetting layer region.

4.2. PHONON SATELLITE OF THE WETTING LAYER STATES

We do not pay attention to the changes of the spectral density in the WL region, which come from the coupling of the WL states to the bound states. In the absence of the quantum dots in the sample, the electron-phonon interaction between the wetting-layer states leads, as it is well known [32], to the formation of the polaron state of an electron, in which the electron is wrapped by the cloud of the optical phonons. The depth of the effective electronic polaron potential hole in the crystal of GaAs may be about 2.5 meV [19]. In this case the spread of the polaron cloud may be estimated as several tens of nanometers. When the density of the quantum dots in the sample is large enough, so that the inter-dot separation is comparable to the size of the polaron, the existence of the polaron states may be not well established. In an overall agreement with the current experiments [4, 14] we shall assume the density of the quantum dots in the sample to be $8 \times 10^{21} \text{ m}^{-3}$. Realizing that the polaronic spectral density peak would be only about several meV below the low-energy edge of the wetting-layer states in GaAs, and having in mind the intention to study a mechanism leading to the appearance of a broad continuous spectral density in the broad range of energy from about $E = -E_{LO}$ until the wetting-layer edge $E = 0$, we shall leave the question of the polaron effect of the carriers in the WL states open and give our attention to that term in the equation (5), which is at $T = 0$ proportional to the electronic population of the wetting layer $N_{\mathbf{k}}$.

That term of the self-energy of the wetting-layer state \mathbf{k} , which is proportional to the wetting-layer population, will be in the lowest order approximation:

$$M_{\mathbf{k}}(E) = \sum_{\mathbf{k}'} \alpha_{\mathbf{k},\mathbf{k}'} \frac{N_{\mathbf{k}'}}{E - E_{\mathbf{k}'} + E_{LO} + i\Delta}. \quad (18)$$

Substituting for $\alpha_{\mathbf{k},\mathbf{k}'}$ we get

$$M_{\mathbf{k}}(E) = \frac{1}{(2\pi)^3} \iiint d^3\mathbf{k}' \frac{p}{|\mathbf{k}' - \mathbf{k}|^2} \frac{N_{\mathbf{k}'}}{E + E_{LO} - E_{\mathbf{k}'} + i\Delta}. \quad (19)$$

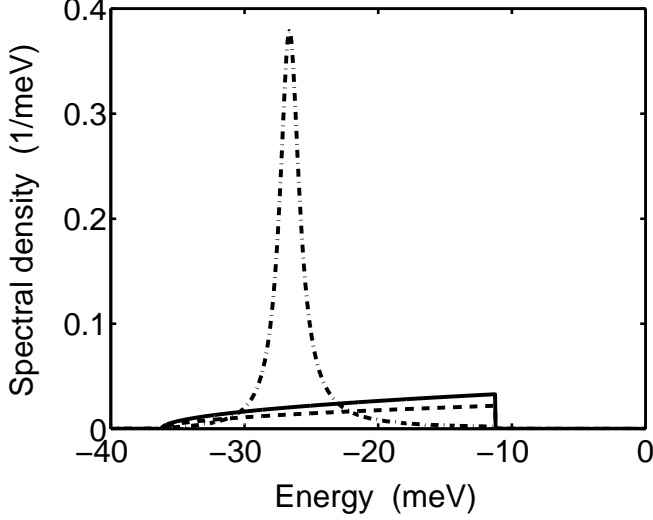


Figure 3. The full and the dash-dot lines give, respectively, the spectral densities of the phonon satellite of the wetting-layer states $\sigma_{tot}(E)$, related to a single quantum dot, and the spectral density $\sigma_n(E)$ of the discrete state of a single dot, both computed for the wetting-layer population $\overline{N}_{\mathbf{k}} = 0.75$. The dashed line is the spectral density of the phonon satellite of the WL states $\sigma_{tot}(E)$ computed for the WL states population $\overline{N}_{\mathbf{k}} = 0.5$.

The integration is extended over the range of $\mathbf{k} < k_m$. Changing the integration variable from \mathbf{k}' to $\mathbf{w} = \mathbf{k}' - \mathbf{k}$, we get

$$M_{\mathbf{k}}(E) = \frac{1}{(2\pi)^3} \iiint d^3\mathbf{w} \frac{p}{|\mathbf{w}|^2} \frac{N_{\mathbf{w}+\mathbf{k}}}{E + E_{LO} - E_{\mathbf{w}+\mathbf{k}} + i\Delta}. \quad (20)$$

When going to the spherical coordinates, the factor $|\mathbf{w}|^2$ in the denominator cancels with the factor coming from the Jacobian of the transformation. In the rest of the integrand we shall make the approximation of "forward scattering", which will consist in neglecting the scattering vector \mathbf{w} in $N_{\mathbf{w}+\mathbf{k}}$ and in $E_{\mathbf{w}+\mathbf{k}}$. The integration limits obviously must obey simultaneously the complicated condition of $|\mathbf{k}| < k_m$ and $|\mathbf{w} + \mathbf{k}| < k_m$. We shall substitute this condition by the condition $|\mathbf{w}| < k_m$ which is obviously exact in the limit of $\mathbf{k} = 0$. The preliminary detailed numerical evaluation of the integral shows, that the simplified evaluation of the integral in (20) is plausible.

The above introduced approximations allow for obtaining the following simple approximate analytical form of the self-energy, namely,

$$M_{\mathbf{k}}(E) = \frac{\gamma}{E + E_{LO} - E_{\mathbf{k}} + i\Delta}, \quad (21)$$

in which $\gamma = pk_m \overline{N_{\mathbf{k}}}/(2\pi^2)$. The distribution of the electrons in all the wetting-layer states \mathbf{k} is constant and equal to $\overline{N_{\mathbf{k}}}$.

The Green's function of the state \mathbf{k} in the wetting layer then is:

$$G_{\mathbf{k}}(E) = \frac{1}{E - E_{\mathbf{k}} - \frac{\gamma}{E + E_{LO} - E_{\mathbf{k}} + i\Delta} + i\Delta}. \quad (22)$$

Looking for the approximate single pole behaviour of this Green's function in the region of energy $E \approx E_{LO}$, we find the following approximate expression of the wetting-layer electron Green's function:

$$G_{\mathbf{k}}(E) = \frac{\gamma/E_{LO}^2}{E + E_{LO} - E_{\mathbf{k}} + i\Delta}. \quad (23)$$

Summing up all the satellite spectral densities of the individual WL states \mathbf{k} within the range of $\mathbf{k} < k_m$ gives the total spectral density $\sigma_{tot}(E)$ of all the extended states. The total spectral density of the phonon satellite of the WL states, related to a single quantum dot, is displayed in the Fig. 3 for the values 0.5 and 0.75 of the population $\overline{N_{\mathbf{k}}}$. In the same graph the single dot bound state spectral density for $\overline{N_{\mathbf{k}}} = 0.75$ is displayed for the purpose of comparing the spectral density of the single dot bound state spectral line with the spectral density of the phonon satellite per one quantum dot.

The total spectral density of the WL satellite, $\sigma_{tot}(E)$, is proportional to the first power of the WL population $\overline{N_{\mathbf{k}}}$, while the integral of the discrete state $\sigma_n(E)$ is constant and equal one. Therefore, increasing the WL population leads to a relative weakening of the discrete state spectral density with respect to the $\sigma_{tot}(E)$. A similar trend is observed in the experimental paper [14], in which the discrete peaks in the sub-wetting layer region of the spectra appear to weaken with respect to the continuous background signal, when the WL population increases.

In the present model of the SAQD sample, the set of the WL states included in the theory is identical with the set of states which are populated with nonzero $N_{\mathbf{k}}$. Only these states contribute then to the formation of the phonon satellite of the WL states. This may lead us to expect, that the experimental spectra of the sub-WL optical response might be influenced also by the distribution function of the population of the WL states.

In the present approach the WL satellite continuum is found in the interval of energy $(-E_{LO}, 0)$, while in the experiment the continuous background may be observed even at the lower energy side of this energy interval. We expect that our model may give the extended range of the continuum upon going to higher orders in the iterative solution of the equations (4,5).

The numerical estimate of the dimensionless quantity γ/E_{LO}^2 gives the value of about 2.8×10^{-2} . The spectral density of the wetting layer satellite

should thus be about 36 times smaller than that of the WL spectral density itself. This number is not easy to compare with the available experimental data, see e. g. [33], because, e. g., we have to be aware of the fact that the optical spectra may be not simply proportional to the spectral densities and also, because our model, assuming the three dimensional wetting layer, may not allow for such a direct comparison of these spectral densities.

5. Conclusions

The present numerical estimate shows that the Fröhlich's mechanism of the electron-phonon coupling in the self-assembled quantum dot samples can contribute considerably to the spectral features observed in the PLE experiments, providing that the population of the electrons in the wetting-layer states is sufficiently large. The optical signal in the sub-wetting layer region of the excitation energy can be expected to have a twofold origin. First, the energy levels of the bound states are broadened by the coupling to the populated WL states. Second, there is a continuous background in the sub-WL region, which is ascribed to the low-energy phonon satellite of the wetting-layer states and which intensity increases linearly with the WL population.

The present conclusions, saying that the electron-LO-phonon coupling can provide the explanation of the continuous features in the optical spectra, do not mean an exclusion of other contributions, like the direct carrier-carrier coupling. These effects remain to be considered in detail, together with the influence of the polaron states and the dimensionality of the WL states.

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